(12) UK Patent Application (19) GB (11) 2 209 164(13) A

(43) Date of A publication 04.05.1989

(21) Application No 8820037.3

- (22) Date of filing 24.08.1988
- (30) Priority data (31) 8720365
- (32) 28.08.1987
- (33) GB

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- (51) INT CL4 C07D 249/18
- (52) UK CL (Edition J) C2C C1452 C220 C365 C662 **U1S** S1349
- (56) Documents cited US 3493539 A Ger. Offen. 3,629,703 (Ciba-Gelgy)
- (58) Field of search UK CL (Edition J) C2C CWD

(54) New benztriazolyl compounds useful as UV-absorbers

(57) A compound of formula I

$$R_1 \xrightarrow{6} \xrightarrow{7} N \xrightarrow{N} N \xrightarrow{R_2} (1)$$

in which R, is selected from $C_{9.32}$ alkyl, $C_{9.32}$ alkenyl, $C_{9.32}$ alkoxy, $-CO-R_4$ and $-(CH_2)_{1.6}-COOC_{6.18}$ alkyl, where R_4 is $C_{8.31}$ alkyl, C₈₋₃₁ alkenyl or C₈₋₃₁ alkoxy; the alkyl group of each significance of R, being uninterrupted or interrupted by one -O-, -S- or -SO₂- group and being unsubstituted or substituted by -N(R_s)₂, OH or halogen, where each R_s, independently, is selected from hydrogen, C₁₋₆ alkyl, C₂₋₆ alkenyl and C₁₋₆ alkoxy;

R₂ has a significance of R₁ independently, of R₁ or is -COOH, hydrogen, hydroxy C_{1.5} alkyl, hydroxy C_{1.5} alkoxy,

-COOC, alkyl, OCOC, alkoxy; Cl, Br, I, OH, C, alkyl, C, alkoxy or C, alkenyl; and R₃ is C_{1.6} alkyl, Cl, Br, I, OH, COOH, hydroxy C_{1.6} alkyl, C_{2.6} alkenyl, hydroxy C_{1.6} alkoxy, COOC_{1.6} alkyl, -OCOC_{1.6} alkoxy or C_{1.22} alkoxy provided that when R₂ contains more than six carbon atoms, R₃ contains six or less carbon atoms.

These compounds are useful as U.V. absorbers in polymeric systems.

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NEW BENZTRIAZOLYL COMPOUNDS

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The invention relates to new benztriazolyl compounds that are useful as U.V. absorbers.

According to the invention, there is provided a compound of formula ${\tt I}$

$$R_1 \xrightarrow{6} \xrightarrow{7} N \xrightarrow{N} N \xrightarrow{R_2} (I)$$

in which R₁ is selected from C₉₋₃₂alkyl, C₉₋₃₂alkenyl,
C₉₋₃₂alkoxy, -CO-R₄ and -(CH₂)₁₋₆-COOC₆₋₁₈alkyl, where R₄ is
C₈₋₃₁alkyl, C₈₋₃₁alkenyl or C₈₋₃₁alkoxy; the alkyl group of each significance of R₁ being uninterrupted or interrupted by one -O-, -S- or -SO₂- group and being unsubstituted or substituted by -N(R₅)₂, OH or halogen, where each R₅, independently, is selected from hydrogen, C₁₋₆alkyl, C₂₋₆alkenyl and C₁₋₆alkoxy;

 R_2 has a significance of R_1 , independently of R_1 , or is -COOH, hydrogen, hydroxy C_{1-6} alkyl, hydroxy C_{1-6} alkoxy, -COOC₁₋₆alkyl, OCOC₁₋₆alkoxy; Cl, Br, I, OH, C_{1-8} alkyl, C_{1-8} alkoxy or C_{2-8} alkenyl; and

 R_3 is $C_{1-6}alkyl,\ Cl,\ Br,\ I,\ OH,\ COOH,\ hydroxyC_{1-6}alkyl,\ C_{2-6}alkenyl,\ hydroxyC_{1-6}alkoxy,\ COOC_{1-6}alkyl,\ -OCOC_{1-6}alkoxy\ or\ C_{1-3\,2}alkoxy\ provided\ that\ when\ R_2\ contains\ more\ than\ six\ carbon$

Case 153-5191 1 - 2 atoms, R3 contains six or less carbon atoms. Preferably, R_1 is in the 5-position as shown in formula I. Any long chain groups present may exist in isomeric form. Compounds of formula I include mixtures thereof. The compounds according to the invention are preferably in 5 liquid form at room temperature (25°C) and normal pressure (1 atmosphere). Preferably, R_1 is R_1' where R_1' is C_{9-22} alkyl, C_{9-22} alkenyl or C_{9-22} alkoxy. More preferably, R_1 is R_1 " where R_1 " is C_{9-18} alkyl. Most 10 preferably R_1 is R_1 "' where R_1 "' is C_{9-14} alkyl. R_1 is especially $C_{12}H_{25}$. Preferably R_2 is R_2 ' where R_2 ' is hydrogen, C_{1-2} alkyl, $C_{2-2,2}$ alkenyl or $C_{1-2,2}$ alkoxy. More preferably R_2 is R_2 " where R_2 " is $C_{1-2\,2}$ alkyl. Most preferably R_2 is R_2 "' where R_2 "' is $C_{6-1\,8}$ alkyl. R_2 15 is especially C_8H_{17} or $C_{12}H_{25}$. Preferably R_3 is R_3 ' where R_3 ' is C_{1-6} alkyl, C_{1-6} alkoxy or C_{2-6} alkenyl. More preferably R_3 is R_3 " where R_3 " is C_{1-6} alkyl, most preferably R_3 is R_3 "' where R_3 "' is C_{1-4} alkyl, especially methyl or t-butyl. Further, according to the invention there is provided a 20 polymeric composition comprising a polymeric material and a compound of formula I. Preferably such a polymeric composition is a lacquer composition based on an acrylic, alkyd, polyester and/or polyurethane resin. Further, according to the invention, there is provided a 25 composition comprising a compound of formula I and a hindered amine light stabiliser, preferably an N-unsubstituted, N-alkyl or N-acyl

substituted 2,2,6,6-tetramethylpiperidine compound.

Preferred hindered light stabilisers are those described in British Published Patent Applications 2,136,805 A; 2,180,537 A and 2,176,482 A and British Patent 2,089,800 B; Belgian Patent 853,476; European Patents 52 579, 52 073 and USP 4,198,334 the contents and preferences of which are incorporated herein by reference.

Compounds of formula I together with a hindered amine light stabiliser in a polymeric composition according to the invention may exhibit synergism in the polymeric composition.

Further, according to the invention, there is provided a method for preparing a compound of formula I comprising reducing and cyclising the compound of formula II

$$R_1 = N - N - R_2$$

$$R_1 = N - R_3$$

$$R_3$$

where the symbols are as defined above, at room or elevated temperature.

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Preferably the reducing and cyclising agent is zinc dust.

The concentration of compound of formula I employed in the polymeric material is suitably 0.01 to 8 % by weight, preferably 0.02 % to 4 % by weight of polymeric material. The compound may be added before, during or after the polymerization step, and may be added in solid form in solution, preferably as a liquid concentrate containing from 20 to 80 % by weight of compound of formula I, or as a solid masterbatch composition containing 20 to 80 % by weight of compound of formula I and 80 to 20 % by weight of a solid polymeric material which is identical with or compatible with the polymeric material to be stabilized. When the compound of formula I is in liquid form it can be used as the liquid without any further

additions.

Suitable polymeric materials include plastic materials for example polyethylene, polypropylene, ethylene/propylene copolymers, polyvinyl chloride, polyester, polyamide, polyurethane,

5 polyacrylontrile, ABS, terpolymers of acrylates, styrene and acrylonitrile, styrene/acrylontrile and styrene/butadiene. Other plastics materials such as polybutylene, polystyrene, chlorinated polyethylene, polycarbonate, polymethylmethacrylate, polyphenylene oxide, polypropylene oxide, polyacetals, phenol/formaldehyde resins and epoxy resins may also be used. Preferred plastic materials are polypropylene, polyethylene, ethylene/propylene copolymers and ABS. Natural polymers for example natural rubber may also be stabilized, as may lubricating oils containing polymeric material.

The compounds of formula I may be incorporated by known methods

15 into the polymeric material to be stabilized. Of particular
importance is blending of the compounds with thermoplastic polymers
in the melt, for example in a melt blender or during the formation of
shaped articles, including foils, films, tubes, containers, bottles,
fibres and foams by extrusion, injection moulding, blow moulding,

20 spinning or wire coating.

It is not essential for the polymeric material to be fully polymerised before mixing with the compound according to the invention. The compounds may be mixed with monomer, prepolymer or precondensate, and the polymerisation or condensation reaction carried out subsequently. This will of course be the preferred method of incorporation of the compounds into thermosetting polymers, which cannot be melt blended.

The compounds of formula I may be used alone or in combination with other stabilizers, for example antioxidants. Examples include sterically hindered phenols, sulphur or phosphorus-containing compounds or mixtures of these. Examples are benzofuran-2-ones, indolin-2-ones and sterically hindered phenols such as beta-(4-hydroxy-3,5-ditert.butylphenyl)-propionyl stearate, methane tetrakis-(methylene-3

(3',5'-ditert.butyl-4-hydroxy-phenyl)propionate), 1,3,3-tris-(2-methyl-4-hydroxy-5-tert.butyl phenyl)butane, 1,3,5-tris (4-tert.butyl-3-hydroxy-2,6-di-methylbenzyl)-1,3,5-triazinyl-2,4,6 (1H, 3H,5H)-trione, bis-(4-tert.butyl-3-hydroxy-2,6-di-methylbenzyl)dithiol-terephthalate, tris (3,5-ditert.butyl-4-hydroxybenzyl) isocyanurate, the triester of beta-(4-hydroxy-3,5-ditert.butylphenyl) propionic acid with 1,3,4-tris-(2-hydroxyethyl)-5-triazinyl-2,4,6 (1H,3H,5H)-trione, bis (3,3-bis-(4'-hydroxy-3'-tert.butylphenyl)-butyric acid) glycol ester, 1,3,5-trimethyl-2,4,6-tris-(3,5-ditert.butyl-4-hydroxy-10-benzyl) benzene, 2,2'-methylene-bis-(4-methyl-6-tert.butylphenyl) terephthalate, 4,4-methylene-bis-(2,6-ditert.-butylphenol), 4,4'-butylidine-bis-(tert.butylmetacresol), 2,2'-methylene-bis- (4-methyl-6-tert.-butyl)-phenol.

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Sulphur containing antioxidative co-stabilizers which may be
used include for example distearylthiodipropionate, di-laurylthiodipropionate, methane tetrakis (methylene-3-hexylthiopropionate),
methane tetrakis (methylene-3-dodecylthiopropionate) and dioctadecyldisulphide. Phosphorus-containing co-stabilizers include for
example trinonylphenyl phosphite, 4,9-distearyl-3,5,8,10-tetraoxadiphosphaspiroundecane, tris-(2,4-ditert.butylphenyl)phosphite and
tetrakis (2,3-ditert.-butylphenyl)-4,4'-biphenylene diphosphonite.
Further additives such as aminoaryl compounds and U.V.-absorbers and
light stabilizers e.g. 2-(2'-hydroxyphenyl-)benzotriazole, 2-hydroxy-benzophenone, 1,3-bis-(2'-hydroxybenzoyl)benzene, salicylates,
cinnamates, benzoates and substituted benzoates, sterically hindered
amines and oxalic acid diamides may be used. Other known types of
additives, e.g. flame retardants and antistatic agents, may also be
added.

The compounds of the invention can also be used in 30 photopolymeric substrates containing photoinitiators for the photopolymerisation.

The compounds of formula I are especially suitable for use in organic polymer-containing coatings, particularly automotive

finishes.

Automotive finishes are generally solutions or dispersions of organic polymers or polymer precursors in organic solvents. The majority are stoving finishes, which require the application of heat, generally above 80°C, in order to harden the finish in an acceptable time once it has been applied to the primer coated metal surface. The hardening step may be accelerated by the use of an acid catalyst. The effect of this heating may be to accelerate the chemical reaction between polymer precursors in a thermosetting system, or to bring about fusion of particles of a thermoplastic polymer.

Many automotive finishes are metallic finishes, which contain flakes of metal, usually aluminium, in order to provide optical effects due to reflection. Such finishes are often two-coat finishes; in which a clear top coat finish is applied over a base coat finish containing a single pigment and/or metal flakes. The compounds of formula I can be in the top coat finish or the ground coat finish, preferably the former. Such two-coat metallic finishes have particular need of U.V.-stabilizers in the top coat, since the polymer in this coat is not protected by light-absorbing pigments, and it is subjected to almost double the normal amount of radiation because of reflection of light from the lower metallic layer.

The compounds of formula I are suitable for use as U.V.-stabilizers in a wide range of liquid finishes, for example those based on combinations of melamine-formaldehyde resins with oil-modified polyester resins, polyacrylate resins with added crosslinkers, or saturated polyesters; or on self-crosslinkers, or saturated polyesters; or on self-crosslinked polyacrylate or polyacrylate resin co-polymerised with styrene.

Further examples are two-component finishes based on an aliphatic or aromatic di-isocyanate and a hydroxy-group-containing polyacrylate, polyester or polyether resin. These polyurethane 2-component finishes are preferably hardened at 60 to 120°C. Thermoplastic polyacrylate resins may also be used, the latter being

particularly useful in metallic finishes, as are also polyacrylate resins with added crosslinkers in combination with melamine-formaldehyde resins etherified with butanol and, further, hydroxy-group-containing polyacrylate resins hardened with aliphatic di-isocyanates. Such polyacrylate resins are described in USP 3,062,753, the contents of which are incorporated herein by reference.

The compounds of formula I are particularly useful in acid catalysed stoving finishes particularly in the top coat of two 10 metallic finishes.

The compounds of formula I may be added to the finish at any stage in its manufacture, and may be added in solid form or in solution, preferably in the form of a liquid concentrate in a suitable solvent or in the form of a dispersion in water or organic solvent or when a liquid, as the liquid.

In practice the compounds of formula I are added to a finish as a solution in organic solvent (as a liquid finish) in which the binder material is between 35 % (low solid finishes) and 70 % by weight (high solid finishes). The binder material of the finish can be in aqueous emulsion or suspension form (as an aqueous finish) in which the binder material part makes up 20 to 30 % by weight. However, the compounds of formula I can be added to known powder finishes.

The compounds of formula I to be added to the liquid or powder finishes before stoving or hardening. Preferably the compounds of formula I are used in liquid finishes or when liquid just as the liquid since it is easy to add exact dosages. It is particularly preferred to use a concentrate (preferably in a hydrocarbon solvent) containing at least 40 % preferably 60 to 80 % by weight of the total weight of the concentrate of a compound of formula I to introduce the compound of formula I to finishes for stoving.

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The addition of from 0.01 to 8 % by weight, preferably 0.2 to 4 % by weight of one or more compounds of formula I gives a clear improvement in the light- and weather-stability of organic pigments in stoving finishes as well as reducing the tendency to hairline 5 cracking and loss of gloss as the result of weathering. This is also found for metallic finishes and excellent long-term stability of the clear top coat of two layer metallic finishes is obtained. In such finishes, the compound of formula I may be added to the metallic undercoat, the clear top coat or both, preferably only to the clear top coat. The metal surface to be finished may be under-coated with primer coatings as is customary in the art of coating metal surfaces.

The invention will now be illustrated by the following Examples in which all parts and percentages are by weight and all temperatures are in degrees Centigrade.

EXAMPLE 1

- a) Preparation of 4-n-dodecyl-2-nitro-2'-hydroxy-5'-methyl-azo benzene;
- 30.6 g of 2-nitro-4n-dodecylaniline are added to 10 ml of water 5 and 35 ml of toluene and then reacted with 25 ml of concentrated hydrochloric acid and then stirred at 50°C for ½ an hour. The temperature is then reduced to -5°C and then the product is diazotised with 7.2 g of sodium nitrite dissolved in 10 ml of water. The reaction is allowed to react for 1 hour whilst adding 50 ml of ethanol. This mixture is slowly added to a mixture of 9.0 g of p-cresol, 12,4 g of NaOH, 300 ml of ethanol and 50 ml of water at 0°C, whilst stirring. After reacting for 3 hours, the reaction mixture is acidified by the addition of conc. HCl solution until the mixture is slightly acid. The aqueous phase is then extracted 3 times with toluene, the organic phases are then added together and then washed neutral. After distilling of the solvent and after chromatographic purification, 19 g of the solid product result (= 54 % of theory), having a melting point of 78-79°C.
- b) Preparation of 5-n-dodecyl-2-[2'-hydroxy-5'-methyl-20 phenyl]-2H-benztriazole.

A solution of 28 g of 4-n-dodecyl-2-nitro-2'-hydroxy-5'methyl-azobenzene (prepared as in part a) above) in 22 ml of water
and 220 ml of isopropanol is reacted with 26 ml of 30 % NaOH and
warmed to 45°C. 20 g of zinc dust is added portionwise over 2 hours.

25 The suspension is stirred at 75°C overnight, cooled and the
isopropanol solution is decanted from the zinc. The organic phase is
then added to 300 ml of water and brought to pH 4 by addition of
concentrated hydrochloric acid. The product is then taken up in 200 g
of hexane and the hexane phase is washed neutral. The solvent is
30 distilled off under vacuum and the product is chromatographically
purified. The resulting product is white.

EXAMPLE 2

a) By a method analogous to that of Example 1a, 24.9 g of 2-nitro-4-n-dodecylaniline can be diazotised and reacted with 18.7 g of 2-sec.-dodecyl-4-methyl-phenol to produce 4-n-dodecyl-2-nitro-3'--sec.-dodecyl-2'-hydroxy-5'-methyl-azobenzene which is a red solid 5 product, having a melting point of 56-58°C.

b) By a method analogous with that of Example 1b, 39 g of 4-n-dodecyl-2-nitro-3'-sec.-dodecyl-2'-hydroxy-5'-methyl azobenzene (produced as in part a) above) is converted to 5-n-dodecyl-

EXAMPLE 3

10 is a orange oil.

Preparation of 5-dodecyl-2-[2'-hydroxy-5'-methyl-3'-octyl-phenyl]-2H benztriazole.

2-[2'-hydroxy-3'-sec.-dodecyl-5'-methyl-phenyl]-2H-benztriazole which

- 20 g of the product of Example 1b is stirred for 6 hours in the presence of 3.3 ml of methane sulphonic acid an excess of octene at 120°C. The product is taken up in hexane, and the hexane phase is washed neutral. After chromatographic purification a non-crystalline oil results which is 5-dodecyl-2-[2'-hydroxy-5'-methyl-3'-octyl-phenyl]-2H benztriazole.
- The 5-dodecyl group in the Example 3 is an isomeric mixture of sec.-dodecyl and n-dodecyl, since isomerisation of the n-dodecyl group occurs to about 50:50.

The octyl group in Example 3 is a mixture of different isomers.

EXAMPLES 4 and 5

By a method analogous with that of Example 1 compounds of the formula

$$R_1$$
 N
 N
 CH_3

in which $R_1\ and\ R_2$ are as in Table 1 below can be prepared from appropriate reactants.

TABLE

Ex. No.	R ₁	R ₂
4	C ₁₂₋₁₄ alkyl (isomer mix)	C ₈ H ₁₇ (isomer mix)
5	C ₁₂₋₁₄ alkyl (isomer mix)	C ₁₂₋₁₄ alkyl (isomer mix)

APPLICATION EXAMPLE A

80 Parts of Viacryl SC 344 (a 50 % solution of an acryl resin from Vianova),

13.9 Parts of Maprenal MF 80 (a 72 % solution of a melamine resin from Hoechst) and

4.1 Parts of Byketol OK (from Byk-Malinckrodt)

is added to 2 parts of the benztriazolyl product of Example 2. After 1 minute the light stabiliser material so formed is dissolved in a

is added to 2 parts of the benztriazolyl product of Example 2. After 1 minute the light stabiliser material so formed is dissolved in a finish. The finish is applied conventionally (according to the known 10 2 layer procedure) to a metallic or single pigment finish whilst still wet by spraying to form a layer having a thickness of 30 to 40 µm. The resulting coating is then hardened at 140° for 30 minutes. The coating shows very good resistance to U.V. light and weathering.

APPLICATION EXAMPLE B

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- 15 A clear finish of
 - 29.5 Parts of Setalux C-1502 XX-60 (a 60 % solution of an acryl resins from Synthese B.V.),
 - 39.2 Parts of Setalux C-1382 BX-45 (a 45 % solution of an acryl resin from Synthese B.V.),
- 21.4 Parts of Setamine US-138 BB-70 (a 70 % solution of a melamine resin from Synthese B.V.),
 - 2.5 Parts of Baysilonoil [(2 % solution in Xylene) from Bayer] and
 - 7.4 Parts of Depanol Y (a solvent from Hoechst)
- 25 is stirred together with 2.5 parts of the benztriazolyl product of Example 2 and 2 parts of an acid catalyst derived from phosphoric acid (Type: Catalyst 269-9 from American Cyanamid) to form a homogeneous mixture. The finish is applied conventionally (according to known 2 layer procedure) to a metallic or single pigment finish

whilst both are still wet by spraying to form a layer having a thickness of 30 to 40 μm . The resulting coating is then hardened at 110° for 20 minutes. The coating shows very good resistance to U.V. light and weathering.

5 APPLICATION EXAMPLE C

A clear finish of

	75	Parts	Macrynal SH 510 N (a hydroxy containing acryl resin from Bayer)
	2	Dants	of Baysilon-oil A [1 % solution in xylene) from
	2	Parts	Of Baysiion-off A (1 % Soldelon in My 14.16) ====
10			Bayer]
	0.3	Parts	of dibutyl zinc dilaurate
	0.35	Parts	diethanolamine
	5.0	Parts	of ethylglycol acetate
	5.0	Parts	of Solvesso 100
15	6.0	Parts	of Xylene and
	6.35	Parts	of butyl acetate

is added to 2.5 parts of the benztriazolyl product of Example 2 and 30 parts of Desmodur N 75 (from Bayer). The homogeneous mixture so formed is applied conventionally (according to the known 2 layer 20 procedure) to a metallic or single pigment finish whilst both are still wet by spraying to form a layer having a thickness of 30 to 40 µm and the resulting coating is hardened over 20 minutes at 80 to 90°. The resulting 2K-PUR coating shows a good resistance to U.V. light and weathering.

25 APPLICATION EXAMPLE D

A single white pigmented finish of

- 14.30 Parts of Setamine US-132 BB70 (a 70 % solution of a melamine resin from Synthese)
- 57.15 Parts of Setal 84 W-70 (a 70 % solution of an alkyd resin from Synthese)
- 5 7.70 Parts of n-butanol
 - 1.85 Parts of butylglycol acetate
 - 9.50 Parts of Xylene and
 - 25 Parts of titanium dioxide (Rutil type)

is added with 1.38 parts of the benztriazolyl product of Example 2.

- 10 The finish is conventionally applied to a grounded steel metal to which a filler of layer thickness 20 to 30 µm has been annealed, by spraying and after standing for 30 minutes at room temperature the steel metal surface is annealed at 120°C for 30 minutes. The resulting coating shows very good resistance to U.V. light and
- 15 weathering. In Application Examples A to D instead of the benztriazolyl product of Example 2, an appropriate amount of the benztriazolyl product of Example 1 or 3, or 4 or 5 can be used.

CLAIMS

1. A compound of formula I

$$R_1 \xrightarrow{6} \xrightarrow{7} N \xrightarrow{N} N \xrightarrow{OH} R_2$$

$$R_3$$

$$(1)$$

in which R₁ is selected from C₉₋₃₂alkyl, C₉₋₃₂alkenyl,
C₉₋₃₂alkoxy, -CO-R₄ and -(CH₂)₁₋₆-COOC₆₋₁₈alkyl, where R₄ is
C₈₋₃₁alkyl, C₈₋₃₁alkenyl or C₈₋₃₁alkoxy; the alkyl group of each
5 significance of R₁ being uninterrupted or interrupted by one -O-, -Sor -SO₂- group and being unsubstituted or substituted by -N(R₅)₂, OHor halogen, where each R₅, independently, is selected from hydrogen,
C₁₋₆alkyl, C₂₋₆alkenyl and C₁₋₆alkoxy;

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 R_2 has a significance of R_1 , independently of R_1 , or is -COOH, hydrogen, hydroxy C_{1-6} alkyl, hydroxy C_{1-6} alkoxy, -COOC₁₋₆alkyl, OCOC₁₋₆alkoxy; Cl, Br, I, OH, C_{1-8} alkyl, C_{1-8} alkoxy or C_{2-8} alkenyl; and

 R_3 is C_{1-6} alkyl, Cl, Br, I, OH, COOH, $hydroxyC_{1-6}$ alkyl, C_{2-6} alkenyl, $hydroxyC_{1-6}$ alkoxy, $COOC_{1-6}$ alkyl, $-OCOC_{1-6}$ alkoxy or C_{1-3} 2alkoxy provided that when R_2 contains more than six carbon atoms, R_3 contains six or less carbon atoms.

- 2. A compound according to Claim 1, in which R_1 is in the 5-position as shown in Claim 1.
- 3. A compound according to Claim 1 or Claim 2, in which R_1' is 20 R_1' where R_1' is C_{9-22} alkyl, C_{9-22} alkenyl or C_{9-22} alkoxy.
 - 4. A compound according to any one of the preceding claims, in which R_1 is R_1 " where R_1 " is C_{9-18} alkyl.

į - 16 -Case 153-5191 A compound according to any one of the preceding claims in 5. which R_1 is R_1''' where R_1''' is C_{9-14} alkyl. A compound according to any one of the preceding claims in 6. which R_2 is R_2 ' where R_2 ' is C_{1-2} alkyl, hydrogen, C_{2-2} alkenyl or 5 C_{1-22} alkoxy. 7. A compound according to any one of the preceding claims in which R_2 is R_2 " where R_2 " is C_{1-2} alkyl. A compound according to any one of the preceding claims in 8. which R_2 is R_2' " where R_2' " is C_{6-18} alkyl. 10 A compound according to anyone of the preceding claims in which R_3 is R_3 ' where R_3 ' is C_{1-6} alkyl, C_{1-6} alkoxy or C_{2-6} alkenyl. 10. A compound according to any one of the preceding claims in which R_3 is R_3 " where R_3 " is C_{1-6} alkyl. 11. A compound according to any one of the preceding claims in 15 which R_3 is R_3''' where R_3''' is C_{1-4} alkyl. 12. A polymeric composition comprising a polymeric material and a compound according to any one of the preceding claims. 13. A polymeric composition according to Claim 12 comprising additionally a hindered amine light stabiliser. 20 14. A polymeric composition according to Claim 12 or Claim 13 which is a lacquer composition based on an acrylic, alkyd, polyester and/or polyurethane resin. 15. A composition according to any one of Claims 12 to 14 in which the amount of benztriazolyl compound of formula I present is 25 from 0.01 to 8 % by weight of the polymeric material.

- 16. A benztriazolyl compound substantially as herein described with reference to any one of Examples 1 to 4.
- 17. A polymeric composition, containing a benztriazolyl compound substantially as herein described with reference to any one 5 of Application Examples A to D.
 - 18. A process for preparing a compound of formula I defined in Claim 1 comprising reducing and cyclising a compound of formula II

$$\begin{array}{c|c}
 & OH & R_2 \\
 & No_2 & R_3
\end{array}$$

where the symbols are defined in Claim 1.

19. A light stabilising composition comprising a compound of 10 formula I defined in any one of claims 1 to 11 and 16 and a hindered amine light stabiliser.

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